

## CLAIMS

1. A porous zeolite shaped body of a zeolite, characterized in that said porous zeolite shaped body is made of a completely  
5 crystallized zeolite composed of tetrapropylammonium ion (TPA) and silica sol in a mixing ratio (TPA/SiO<sub>2</sub>) of 0.015 to 0.08 by mole.
2. A porous zeolite shaped body of a zeolite, characterized in that the porous zeolite shaped body is made of a zeolite still under crystallization and composed of tetrapropylammonium ion (TPA) and  
10 silica sol in a mixing ratio (TPA/SiO<sub>2</sub>) of 0.02 to 0.12 by mole.
3. A zeolite intermediate body, characterized in that the zeolite shaped body as claimed in Claim 1 or Claim 2 contains further a template, and a template-containing zeolite membrane having a composition the same as or similar to that of the shaped body is  
15 formed thereon.
4. A zeolite layered composite comprising said zeolite shaped body and said zeolite membrane layered thereon, characterized in that the composite is produced by removing said template from said zeolite shaped body and said template-containing zeolite membrane by  
20 calcining the zeolite layered intermediate body as claimed in Claim 3.
5. A method for producing a zeolite layered composite, characterized by layering a template-containing zeolite membrane having a composition the same as or similar to that of a zeolite  
25 shaped body of a zeolite still under crystallization and composed of tetrapropylammonium ion (TPA) and silica sol in a mixing ratio

(TPA/SiO<sub>2</sub>) of 0.015 to 0.08 by mole and containing a template therein on said zeolite shaped body, and simultaneously removing the template from said zeolite membrane and said zeolite shaped body by calcining the resulting layered product to obtain a zeolite layered composite comprising said zeolite shaped body and said zeolite membrane layered thereon.

6. A method for producing a zeolite layered composite, characterized by layering a template-containing zeolite membrane having a composition the same as or similar to that of a zeolite shaped body of a zeolite still under crystallization and composed of tetrapropylammonium ion (TPA) and silica sol in a mixing ratio (TPA/SiO<sub>2</sub>) of 0.02 to 0.12 by mole and containing a template therein on said zeolite shaped body, and simultaneously removing a template from said zeolite membrane and said zeolite shaped body by calcining the resulting layered product to obtain a zeolite layered composite comprising said zeolite shaped body and said zeolite membrane layered thereon.

7. A porous zeolite shaped body of a zeolite, characterized in that the porous zeolite shaped body has an average particle diameter of 1.0  $\mu\text{m}$  or larger, a bending strength of 1.5 MPa or higher, and a difference in pressure between a feed side and a permeation side of 1.0 atmospheric pressure or lower at 10 ml/cm<sup>2</sup> · min of helium gas permeation flux when a thickness of the porous zeolite shaped body is adjusted to be 1.8 mm

8. A zeolite layered intermediate body, characterized in that the zeolite shaped body as claimed in Claim 7 contains, further a template

and a template-containing zeolite membrane having a composition the same as or similar to that of the shaped body is layered thereon.

9. A zeolite layered composite comprising said zeolite shaped body and said zeolite membrane layered thereon, characterized in that  
5 the zeolite layered composite is formed by removing said template from said zeolite shaped body and said template-containing zeolite membrane by calcining the zeolite layered intermediate body as claimed in Claim 8.

10. A method for producing a zeolite shaped body, characterized  
10 by adding a tetrapropylammonium hydroxide (TPAOH) solution and tetrapropylammonium bromide (TPABr) to silica sol in such a manner that mixing ratios  $[\text{TPAOH}/(\text{TPAOH} + \text{TPABr})$  and  $\text{TPABr}/(\text{TPAOH} + \text{TPABr})]$  of tetrapropylammonium hydroxide (TPAOH) and tetrapropylammonium bromide (TPABr) to a total amount of tetrapropylammonium ion (TPA)  
15 become 0 to 99% by mole and 1 to 100% by mole, respectively to prepare a solution, drying thus prepared solution by kneading the solution, shaping thus obtained dried gel, and subjecting thus shaped body to crystallization treatment.

11. A method for producing a zeolite shaped body, characterized  
20 by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol to prepare a solution, spraying thus prepared solution to dry, shaping thus obtained dried gel, and subjecting thus shaped body to crystallization treatment.

12. A method for producing a zeolite layered intermediate body,  
25 characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution and tetrapropylammonium bromide (TPABr) to silica sol in

- such a manner that mixing ratios  $[\text{TPAOH}/(\text{TPAOH} + \text{TPABr})]$  and  $\text{TPABr}/(\text{TPAOH} + \text{TPABr})$  of tetrapropylammonium hydroxide (TPAOH) and tetrapropylammonium bromide (TPABr) to a total amount of tetrapropylammonium ion (TPA) become 0 to 99% by mole and 1 to 100%,  
5 respectively to prepare a solution, drying thus prepared solution by kneading the solution, shaping thus obtained dried gel, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to said prepared solution,  
10 and forming a template-containing zeolite membrane on said zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-containing zeolite membrane.
13. A method for producing a zeolite layered intermediate body,  
15 characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol, spraying thus prepared solution to dry, shaping the obtained dried gel, subjecting the shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution having the same or similar  
20 composition as or to that of said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-containing zeolite membrane.
- 25 14. A method for producing a zeolite layered composite, characterized by adding a tetrapropylammonium hydroxide (TPAOH)

solution and tetrapropylammonium bromide (TPABr) to silica sol in such a manner that the mole ratio of mixing ratios [TPAOH/(TPAOH + TPABr) and TPABr/(TPAOH + TPABr)] of tetrapropylammonium hydroxide (TPAOH) and tetrapropylammonium bromide (TPABr) to a total amount of tetrapropylammonium ion (TPA) become 0 to 99% and 1 to 100%, respectively to prepare a solution, drying thus prepared solution by kneading the solution, shaping thus obtained dried gel, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said prepared solution, forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-containing zeolite membrane, and then calcining the layered body to simultaneously removing the template.

15. A method for producing a zeolite layered composite, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol to prepare a solution, spraying thus prepared solution to dry, shaping thus obtained dried gel, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said prepared solution, forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising said zeolite shaped body and said template-

containing zeolite membrane, and then calcining the layered body to simultaneously removing the template.

16. A porous zeolite shaped body of a zeolite, characterized in that area of parts (sound parts) where respective particles are  
5 clearly observed by grain boundary fracture among particles composing the zeolite shaped body in microstructure observation of the fractured surface of the shaped body occupies 70% or more in the entire area of the fractured surface.

17. A zeolite layered intermediate body, characterized in that the  
10 zeolite shaped body as claimed in Claim 16 further contains a template, and a template-containing zeolite membrane having a composition same as or similar to that of the shaped body is formed on the shaped body.

18. A zeolite layered composite comprising a zeolite shaped body  
15 and a zeolite membrane formed thereon, characterized in that the zeolite layered composite is produced by removing said template from said zeolite shaped body and said template-containing zeolite membrane by calcining the zeolite layered intermediate body as claimed in Claim 17.

19. A method for producing a zeolite shaped body, characterized  
20 by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 to prepare a solution, drying thus prepared solution by kneading  
25 the solution, wet pulverizing thus obtained dried gel, spraying thus obtained slurry to dry, shaping thus obtained dried granular

substance, and subjecting thus obtained substance to crystallization treatment to obtain a zeolite shaped body.

20. A method for producing a zeolite shaped body, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of  
5 tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 to prepare a solution, spraying thus prepared solution to dry, shaping thus obtained dried gel, and subjecting thus obtained gel to crystallization treatment to obtain a zeolite shaped body.
- 10 21. A method for producing a zeolite layered intermediate body, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 to prepare a solution, drying thus prepared solution by  
15 kneading the solution, wet pulverizing thus obtained dried gel, spraying thus obtained slurry to dry, shaping thus obtained dried granular substance, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution having the same or similar  
20 composition as or to said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising the zeolite shaped body and the template-containing zeolite membrane.
- 25 22. A method for producing a zeolite layered intermediate body characterized by adding a tetrapropylammonium hydroxide (TPAOH)

solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 by mole to prepare a solution, spraying thus prepared solution to dry, shaping the obtained dried gel, subjecting to thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising a zeolite shaped body and a template-containing zeolite membrane.

23. A method for producing a zeolite layered composite, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 to prepare a solution, drying thus prepared solution by kneading the solution, wet pulverizing thus obtained dried gel, spraying thus obtained slurry to dry, shaping thus obtained dried granular substance, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body, immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said prepared solution, and forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising a zeolite shaped body and a template-containing zeolite



membrane, and then simultaneously removing a template by calcining the layered body.

24. A method for producing a zeolite layered composite, characterized by adding a tetrapropylammonium hydroxide (TPAOH) solution to silica sol in such a manner that a mixing ratio (TPA/SiO<sub>2</sub>) of tetrapropylammonium ion (TPA) to said silica sol becomes 0.015 to 0.08 to prepare a solution, spraying thus prepared solution to dry, shaping thus obtained dried gel, subjecting thus shaped product to crystallization treatment to obtain a zeolite shaped body,
- 10 immersing said zeolite shaped body in a solution with the same or similar composition as or to that of said prepared solution, forming a template-containing zeolite membrane on the zeolite shaped body by hydrothermally synthesizing it thereon to produce a layered body comprising a zeolite shaped body and a template-containing zeolite
- 15 membrane, and then simultaneously removing a template by calcining the layered body.